REMARKS

Claims 1-10 have been rejected. Applicant notes with appreciation the withdrawal of the previous grounds of rejections. As will be discussed below, it is believed that Claims 1-10 are also patentable over the new art cited since none of the cited patents disclose or suggest the use of an electron bombardment ion source for a neutron generator, as presently set forth in Claim 1.

Claims 1, 3-8, and 10 have been rejected under 35 U.S.C. §102(b) as being anticipated by Bernardet (U.S. 5,215,703). In paragraph 2 of the Office Action, the Examiner states "Bernadet discloses a high flux neutron generator comprising an electron bombardment ion source..." Although the ion sources described by Bernadet may use the electron collision (bombardment) physical phenomenon to generate ions, none of the ion sources described in the embodiments are "electron bombardment" ion sources as presently claimed. Bernadet discloses the following embodiments:

1. Column 3, line 42: "According to a first embodiment using magnetic confinement, the ion source is formed by at least one elementary source of the Penning structure."

2. Column 3, line 61: "According to a second embodiment also using magnetic confinement, the ion source is formed by a structure of the inverted magnetron type."

3. Column 4, line 16: "According to a third embodiment using electrostatic confinement, the ion source is of the orbitron type comprising..."

4. Column 4, line 22: "According to a fourth embodiment also using electrostatic confinement, the ion source is of the electrostatic Reflex type (SIRE) and..."

Those skilled in the art of ion beam devices follow text book definitions such as included here as Exhibit A, F.A. White, Mass Spectrometry in Science and Technology, John Wiley & Sons, 1968. As listed in the table of contents, the electron bombardment source is treated as a unique method of ion production, whereas the Penning source, p. 70, is treated as a subset of discharge (plasma) sources. The electron bombardment source is described on p. 58, "No other source provides a comparable stability", which is a required characteristic for propagating an ion beam down a small diameter needle as required for tumor treatment, as presently proposed. Further, in Fig. 3.1 p. 59, White illustrates the electron bombardment source using a small diameter electron beam derived from a filament creating ions in a small volume by electron collisions with gas molecules and therefore resultant ions can be focused with great precision.

In contrast thereto, White shows in Fig. 3.7, p. 70, the Penning discharge source (also described by Reifenshweiler, Neutrons from Small Tubes-Philipps Tube:

Continuous or Pulsed Operation, Nucleonics, Vol. 18, No. 12, Dec. 1960) contains a large ionization volume that is efficient for producing high ion currents, but with a large half angle of divergence of ions leaving the source that is difficult to propagate down a narrow needle. Also, White describes in par. 2, p. 61, that "currents of 10^{-7} to 10^{-14} amperes are monitored in typical analyzers". The present claimed invention employs modified ion optics of the electron bombardment source components - namely providing an ion exit slit and focusing apertures of 3mm or greater which may achieve ion currents of $> 10^{-5}$ amperes that is 100 times higher than described by White and therefore can be described as a 'high current electron bombardment source'. The claims of this invention are based upon such a high current electron bombardment source suitable for propagation of an ion beam down a narrow needle with a thermonuclear reaction target at its end for neutron generation use in brachytherapy tumor treatment. Up to the present invention, discharge (plasma) sources have been used almost exclusively for neutron generators because they deliver high ion currents over large target areas.

The Examiner contends that Bernadet discloses an ion bombardment source in the embodiment of Fig. 7. However, Fig. 7 refers to the orbitron structure embodiment noted at column 4, line 16. At column 7, line 61, the patent states "The operating principle is as follows:...and thus oscillate in the interior of the structure for a longer period, by which the ionization probability is strongly increased and a discharge with the formation of a plasma is created." Thus this

embodiment is a discharge (plasma) source not electron bombardment.

In the sentence bridging pages 2 and 3 of the Office Action, the Examiner further states: "This focusing of the ions by means of the acceleration electrode configuration and the applied voltage on these electrodes constitute the rasterizing means." Again, there is a definition problem here. Focusing of light or ions is associated with achieving the intended size and definition of the beam at its focal plane, and does not provide for movement of the beam. Rasterizing generally means movement of the beam along the focal plane in a controlled pattern, e.g., cathode ray tube used in televisions.

Claims 2 and 9 have been rejected under 35 U.S.C. §103(a) as being unpatentable over Bernardet in view of Reifenschweiler. As noted above, neither Bernadet nor Reifenschweiler disclose an electron bombardment ion source. Instead they each have the use of a gas plasma or Penning discharge source. Reifenschweiler refers many times to the source as a Penning discharge source, not an electron bombardment ion source.

Applicant has developed a high ion current electron bombardment source based neutron generator to provide a useful dose in brachytherapy treatment of cancerous tumors. The definition of electron bombardment source in the attached sections of the textbook by White differentiates this type of ion source from others as they have been defined. Thus, electron collisions with gas molecules may be the basis for ion production in the Penning discharge source, but this has been defined as a discharge source. Put simply, nowhere is there any suggestion of the use of

standard electron bombardment ion source for use in a neutron generator, as presently claimed.

In view of the foregoing, reconsideration and withdrawal of the rejection, and allowance of the claims at an early date are earnestly solicited.

Respectfully submitted,

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I hereby certify that this correspondence is being deposited with the United States Postal Service as first-class mail in an envelope addressed to: Commissioner of Patents, P.O. Box 1450, Alexandria, VA 22313-1450 on July 31, 2003.

By: _____ Dated: July 31, 2003
Thomas M. Galgano, Esq.

Mass Spectrometry in Science and Technology

. A. White

HN WILEY & SONS, INC., NEW YORK · LONDON · SYDNEY

1968

Chapter 3

Methods of Ion Production

In a given experimental situation an ion source may represent either the simplest or the most complex component of a spectrometer. In most instances, however, the production of a suitable beam of ions is far from a trivial assignment, and as additional applications are found for mass spectrometry, the problems of source technology can also be expected to increase. The preparation of biological samples will call for different techniques from those appropriate to semiconductors, and gas analysis methods cannot be expected to apply to the investigation of surface monolayers of metals. The choice of a source will also be severely conditioned by the total sample size.

General criteria, however, can be listed if consideration is given to the general kinematics of ion trajectories and to the simple equation relating to the image "line width" at the detector focal plane. For a symmetric magnetic lens, a simple relationship exists between the object slit width, S_o, the half angle of rays emerging from the defining slit, α, the potential through which ions have been accelerated, V, the spread in ion energy corresponding to a potential difference, ΔV, the line width, W (at the image plane), and the radius of curvature, R, of the magnetic sector [1]. The relationship is

$$W = S_o + R\left(\alpha^2 + \frac{\Delta V}{V}\right). \tag{3.1}$$

If α is expressed in radians, a substitution of values for R, S_o, and $\Delta V/V$ will yield an approximate value for the image width. Equation (3.1) and other general considerations suggest the generally desirable properties of ion sources and ion beams:

1. The supply of ions should be sufficiently intense to be compatible with the analyzer geometry and detector sensitivity.
2. The beam should have an energy spread that is small compared with the total accelerating voltage.

3. The ion source slit width should be small compared with the radius of curvature of the analyzer.

4. The half-angle of divergence should be commensurate with the image line width—as expressed in (3.1) (unless special magnetic pole-shaping of the analyzer is employed which can minimize the $\alpha^2 R$ contribution).

5. If the sample to be analyzed is very small, it is important that a high percentage of sample atoms become ionized.

6. Ideally, the source should be selective against unwanted ions that might appear in the same portion of the mass spectrum.

7. The source should not have a "memory" that would yield ions from the contamination of prior analyses.

8. Ion emission should be reasonable stable with time.

9. The source should minimize the chemical procedures required prior to mass spectral analysis.

Other considerations relate to beam spreading caused by coulomb repulsion (for intense beams), molecular dissociation, charge exchange, and mass discrimination. In most instances, of course, the prerequisite characteristics of the source depend upon the experimental objectives. A mass spectrometric study, for example, might be primarily concerned with the magnitude of an ion beam energy spread. Furthermore, there are situations in which a multiplicity of ions of the same mass but in several charge states provides a more positive elemental identification than can be obtained from a single atomic group.

The most important ion sources in modern mass spectrometry are reviewed below. They include electron bombardment, surface ionization, arcs, vacuum spark, ion impact, and field emission. Photoionization and laser sources are also mentioned, as they are becoming of interest in special applications.

ELECTRON BOMBARDMENT

The production of positive ions by electron impact is a technique universally employed for general gas analyses. With a suitably designed source, one can generate a copious beam of ions having a reasonably small energy spread. Furthermore, the ion beam intensity can be well controlled because the ionizing electron beam is generally space-charge limited. No other source provides a comparable stability in ion production. It is a preferred type for general analytical work as it can be utilized for nearly all gases, volatile compounds, and metallic vapors. Another important advantage of this source is that the ionization of a

complex molecule can be controlled by varying the kinetic energy of the bombarding electrons. Both the number and specie of ion fragments can be altered in a manner so as to yield important data relating to the structural formula of the gas that is being subjected to mass spectral assay.

Gas inlet systems, ovens, valves, and gas mixing reservoirs are usually designed according to specific applications. Gas pressures in the ionizing region may vary from 10^{-2} to 10^{-5} torr or lower. Substances having a high vapor pressure at room temperature may be introduced either directly or indirectly into the source region, and the evaporation of inorganic solids from high temperature crucibles is a standard technique. The electron bombardment source itself, however, always possesses the common elements of (a) an electron-producing filament and electron trap, (b) high-voltage electrodes for accelerating the positive ions generated by electron impact, and (c) collimating slits to provide beam definition. A more complex source that provides additional focusing electrodes and which is typical of many gas sources is shown in Figure 3.1.

Electrons are emitted from the filament by thermionic emission and traverse the ionization chamber region. As ions are formed, they are accelerated by a drawing-out electrode, subjected to electrostatic potentials of focusing plates, and further accelerated through a potential of several kilovolts. Collimating slits effectively determine the divergence angle of the beam that enters the main section of the mass spectrometer, and beam-centering plates are also sometimes employed to increase the ion beam transmission.

It might be presumed that the energy of the ionizing electrons need

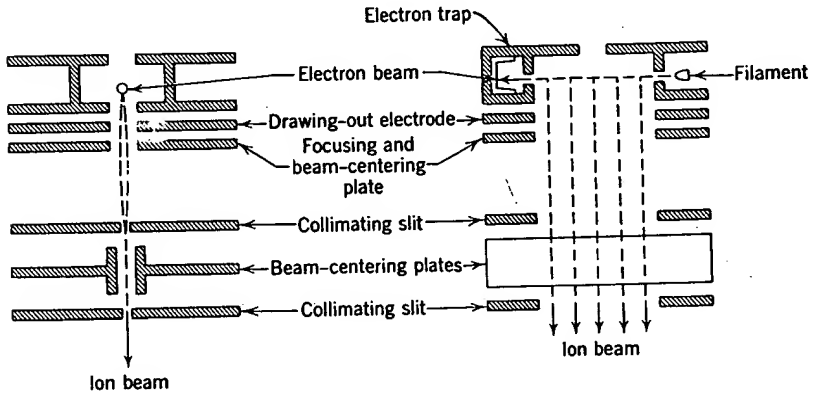

Fig. 3.1 Schematic diagram of an electron-bombardment source.

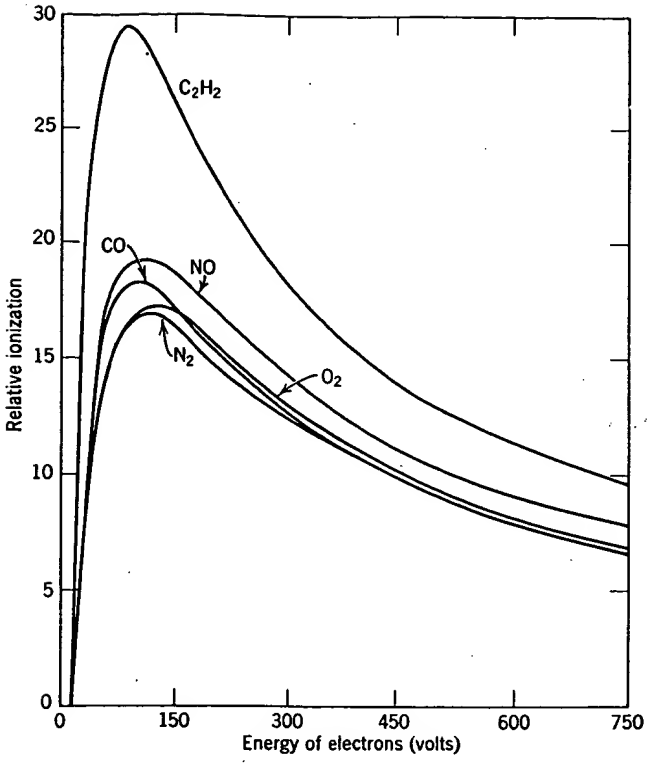

Fig. 3.2 Probability of ionization as a function of electron energy [2].

only exceed the first ionization potential of the sample gas. For achieving a maximum ionization efficiency, however, the electron beam energy must always substantially exceed this value; Figure 3.2 indicates the relative yield of ions produced as a function of electron energy [2].

It will be noted from Figure 3.2 that although there exist large differences in the positive ion yield of various gases, most ionization maxima occur in a region of 50–100 eV, and a practical operating voltage for the electron beam is about 75 V. The number of positive ions produced by the electron beam is directly proportional to the density of gas molecules at pressures $<10^{-4}$ torr. At higher pressures, the production of ions is not linearly related to gas density, owing to space-charge effects and recombination phenomena. The bombarding electrons will also produce atomic ions and molecules in many excited states. Atomic species will lose energy by radiation or by energy exchange in collisions. Molecules, however, can also undergo de-excitation by dissociation, giving

as well as ions of the parent molecule.

In a typical source, the electron beam is orthogonal to the trajectory of the ions, and the region from which ions are drawn into the accelerating region is small. Tungsten and rhenium filaments supply a copious electron current (of order of magnitude $\sim$1 mA) and this current is stabilized by appropriate circuitry in order to obtain a constant ion beam intensity. Electron beam stability may approach 1 part in 10^4, although this condition rarely results in a comparable stability with respect to the ion beam. The magnitude of the ion beam current will depend on several parameters—the specific gas, pressure, size of collimating slits, etc., in addition to the electron-bombarding current. Currents of 10^{-7} to 10^{-14} A are monitored in typical analyzers. The spread in energy of ions produced by typical electron-bombardment sources may be between 10 and 50 eV, but special techniques can reduce this value. For high-resolution work, however, a double-focusing system—or some type of energy discrimination—is essential.

The sensitivity of this type of source varies over many orders of magnitudes. Milligram amounts of sample may be needed, depending on the requirements of precision and other factors. The minimum gas sample is restricted primarily by the background spectrum of residual gases, the pumping system, and the detector sensitivity. Reynolds [3], using a small analyzing tube and employing a static vacuum, was able to detect xenon and argon with only $\sim 10^7$ atoms of sample. Such measurements, however, are quite beyond the capabilities of conventional analytical instruments. In fact, background gases often seriously limit the detection of trace gases because of the "memory" that an ion source retains for occluded gases on walls and surfaces. Despite such difficulties, detection limits of 1 part in 10^7 for trace gases have often been observed.

Precision values ± 0.1 to $\pm 0.01\%$ in isotopic abundance ratios can be achieved, but few investigators claim that absolute accuracies of conventional analyses approach the latter value. Utilization of standards, statistical methods, and knowledge of instrumental biases are prerequisites for achieving best values, even with an electron-bombardment type source.

Negative as well as positive ions can, in principle, be generated by electron-impact phenomena for compound species (e.g., $XY + e \rightarrow X^+ + Y^- + e$). Electron-capture processes present an alternative mode of formation. But, to date, mass spectral analysis of negative ions has been restricted to a few special problems, and detailed research studies.

Many electron-bombardment sources have been designed and been

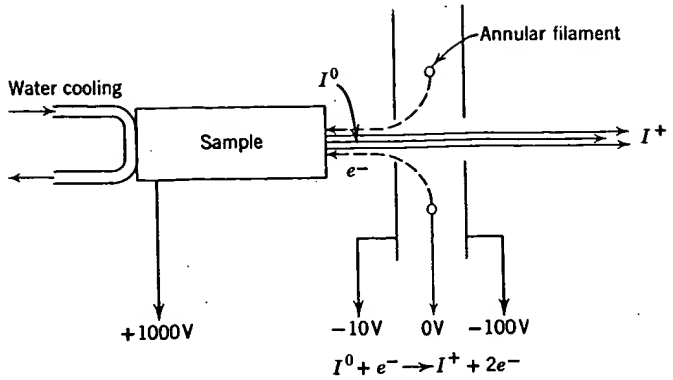

Fig. 3.3 Electron-bombardment source proposed by Honig [4].

successfully applied to the analysis of gases or metallic vapors. One specialized type of electron bombardment source has recently been proposed by Honig [4] for vaporizing refractory solids. The basic concept concerns having a single electron beam perform the dual function of vaporization and ionization—as indicated in Figure 3.3. A fraction of the neutral species evaporated by the heat of electron impact are subsequently ionized and drawn out axially into the mass spectrometer. This scheme will generate ions over a large energy range, thus requiring a double-focusing system. Nevertheless, the proposed system has merit on the basis of simplicity, and it may prove to be useful for the analysis of special samples that are difficult to analyze by more conventional methods.

SURFACE IONIZATION

The phenomenon of surface or thermal ionization has been utilized as a basis for a second and increasingly important ion source in mass spectrometry. Consider an atom or molecule that is evaporated from a heated surface. At sufficiently elevated temperatures, the emission of neutral vapor will be accompanied by positive ions (i.e., a fraction of the atoms or molecules will escape from the surface in an electron-deficient state). In such an ionization mechanism the hot metal surface is said to have a higher affinity for retaining an orbital electron of an escaping atom than the atom itself. The quantitative relationship for predicting the ratio of ionized to neutral atoms was first suggested

the detector providing the mass spectrometer has adequate resolution. The ΔV can be adjusted to about 10 V, but it should not exceed the first ionization potential of residual gases in the source region as in such a case an electron impact spectra would be superposed upon the metal spectrum. It will also be noted that if the ionization filament is operated at a very high temperature (regardless of sample emission rate), a larger number of ions will leave the ionization filament as atoms rather than as molecules that tend to be dissociated. Thus a less complex mass spectrum results.

The parallel triple filament [11] is another multiple-filament type reported to be useful in comparing an unknown sample with a standard. Some increase in ionization efficiency is also claimed because of the better solid angle and the probability of multiple collisions on the part of neutral atoms or molecules.

Investigators have also explored various possibilities for enhancing the ion emission from hot filaments by effectively changing the work function of the surface or by treating the source with a reducing agent [12]. Such techniques have proved to be highly successful in specific instances when the ion emission problem can be restricted to a particular element or metallic compound. The surface-ionization source does not possess the stability of emission of the electron bombardment type, but it has been applied routinely for sample assay in the 10^{-9}-gm range. Under favorable conditions 10^{-14} gm of sample can be detected.

DISCHARGE SOURCES

An important class of mass spectrometer sources includes specific types that utilize discharge phenomena for production of ions. Considerable literature is available reviewing this field in detail. Therefore this book's discussion will be limited to the most important characteristics of sources generally categorized as (a) gaseous discharges, (b) low-voltage arcs, (c) sparks, and (d) the vibrating arc.

Gaseous Discharges

The production of ions by gaseous discharge holds more historical than contemporary interest. Gaseous discharge techniques were employed by almost all the early mass spectroscopists—including Thomson, Aston, and Bainbridge. Ions were produced in a "glow discharge" tube in which cathode-anode voltages ranged from 10,000 to 50,000 V. Because ions are produced at intermediate pressures, differential pumping was usually employed to the ion tube and analyzer regions. Extraction of ions was through an aperture in the cathode, the cathode thus having a dual

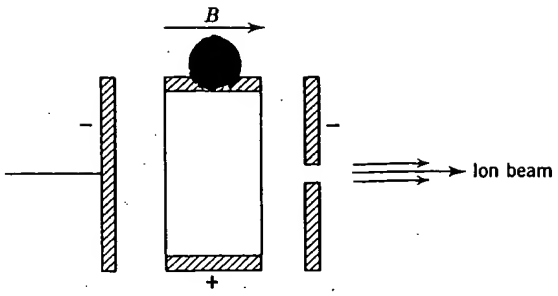

Fig. 3.7 Cold-cathode ionization source devised by Penning.

Any electrons that are present will undergo oscillating trajectories through the anode. The magnetic field lines constrain the electron paths to tight helices, enhancing the degree of ionization. At pressures of 10^{-3} to 10^{-4} torr and with interelectrode potentials of several kilovolts, a self-sustaining discharge can be maintained without a hot filament to supply electrons. This permits operation of the ion source at gas pressures about two orders of magnitude below that of the conventional type. If the anode and cathode are arranged to be perpendicular rather than collinear to the ion beam that is drawn into the mass spectrometer, the effective ion energy spread can be reduced to about 25 eV.

This same general method has been employed (but with auxiliary thermionic emission as noted below) for the production of ions in isotope separators. Ion currents up to 0.1 mA have been achieved and a high yield ($\sim$10% ions to sample atoms consumed) has been reported [14].

Low-Voltage Arcs

Low-voltage arcs in a variety of geometries have been developed for mass spectrometers, cylotrons, and isotope separators. The basic elements are shown in Figure 3.8. The essential distinction between the gaseous discharge and arc source is that the latter is not self-sustaining, but requires a hot filament to furnish a copious supply of ionizing electrons to maintain the arc. The electron beam is at right angles to the direction from which ions are extracted from the arc plasma. Improved electron-beam collimation can be achieved by means of auxiliary magnets having field lines collinear with the electron trajectories. Such a source has been termed a "duoplasmatron." While the initial discharge must usually be triggered by the introduction of a "buffer gas" or high electrode potentials, low voltages across the arc can be maintained at quite low pressures. Using a duoplasmatron, Koch [15] has reported the production